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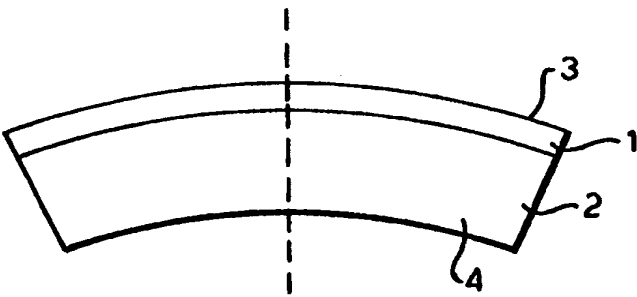
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| (54) Title: MULTIPLE LAYER MONOLITHIC STRUCTURE AND USE THEREOF  (57) Abstract A multiple layer monolithic structure comprising at least a first layer of porous material having a relatively high number of pores per cm and a second layer of porous material having a relatively low number of pores per cm, a catalyst or catalyst precursor comprising the multiple layer monolithic structure, wherein at least the first layer of the monolithic structure has been provided with a catalytically active material or a precursor therefor, and a process for the catalytic conversion of a fluid substrate comprising contacting the substrate with the catalyst as defined, in particular a process for the catalytic partial oxidation of a hydrocarbonaceous feedstock. | | |

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MULTIPLE LAYER MONOLITHIC STRUCTURE AND USE THEREOF

The present invention relates to a multiple layer monolithic structure, to catalysts or catalyst precursors comprising such a structure provided with one or more catalytically active materials and to processes in which the structure is used as catalytic support, in particular conversion processes, more particularly the preparation of carbon monoxide and hydrogen by the partial oxidation of a hydrocarbonaceous feed, nitric oxide reduction processes, and ethylene oxidation.

Monolithic structures such as ceramic foams are known for various applications, in particular more recently as supports for catalytically active materials fulfilling several requirements simultaneously, as described in "Ceramic foam catalyst supports: Preparation and properties" by JT Richardson and MV Twigg, published in Mat. Res. Soc. Symp. Proc. 368 (1995), p. 315-320. Open pore ceramic foams may be made from materials with high temperature resistance, and promote surface-catalysed reaction by means of tortuous flow patterns by virtue of connecting adjacent pores or "cells" providing non-linear channels. Open pore ceramic foams enable the passage of gases at high space velocities and acceptable pressure drop, are readily shaped and provide good conductivity.

European Patent Application EP-A-0 303 438 discloses a process for the catalytic partial oxidation of a hydrocarbonaceous feedstock in which a gaseous mixture of the hydrocarbonaceous feedstock, oxygen or an oxygen-containing gas and, optionally, steam, is introduced into a catalytic partial oxidation zone to contact a catalyst retained therein, which catalyst has a surface area to

volume ratio of at least $5 \text{ cm}^2/\text{cm}^3$. The catalyst may be of a variety of forms, for example sheets of corrugated metal packed to form elongate channels therethrough or wire mesh. However, preference is given in EP-A-0 303 438 to the use of catalysts in the form of extruded honeycomb monoliths. These monoliths comprise a large number of parallel channels extending through the structure in the direction of flow of the feed and product gases.

D.A. Hickman and L.D. Schmidt ("Synthesis Gas Formation by Direct Oxidation of Methane over Pt Monoliths", Journal of Catalysis 138 (1992), p. 267-282) have conducted experiments into the partial oxidation of methane in the presence of catalysts comprising either platinum or rhodium as catalytically active material. The catalysts employed were in the form of metal gauzes, metal-coated foam monoliths or metal-coated extruded monoliths. The foam monoliths were of alpha-alumina and described as having an open cellular, sponge-like structure. The samples employed had 30 to 50 pores per inch (ppi). The extruded monoliths were cordierite extruded monoliths, having 400 square cells/in² (20 ppi) and consisted of straight parallel channels giving laminar flows of gases through the channels under the conditions of gas flow rate studied.

J.K. Hockmuth ("Catalytic Partial Oxidation of Methane over a monolith Supported Catalyst", Applied Catalysis B: Environmental 1 (1992), p. 89-100) reports the catalytic partial oxidation of methane using a catalyst comprising a combination of platinum and palladium supported on a cordierite monolith body having 46.5 cells/cm² (17 ppi).

The monoliths used in the above-mentioned references are either in the form of a honeycomb structure or in the form of a foam containing a relatively low number of

pores (at most 50 ppi), i.e. pores having a relatively large diameter. This will result in a sufficiently low pressure drop across the monolith structure to allow very high space velocities. However, in catalytic partial oxidation processes, structures having straight channels or having relatively large pores may result in only moderate hydrocarbon conversion rates and selectivity, due to the occurrence of gas phase reactions. Especially at the high temperatures at which catalytic partial oxidation processes are carried out, gas phase reactions easily occur.

The specification of European patent application EP-A-0 656 317 contains a description of a process for the catalytic partial oxidation of a hydrocarbon feedstock in which the hydrocarbon is mixed with an oxygen-containing gas and contacted with a catalyst. The catalyst is retained in a fixed arrangement having tortuosity of at least 1.1 and having at least 750 pores per square centimetre (70 ppi). The fixed bed arrangement in which the catalyst is present is in the form of particles or as a monolith structure, such as a ceramic foam. The catalyst preferably comprises a catalytically active metal supported on a carrier. Suitable carrier materials are described as including the refractory oxides, such as silica, alumina, titania, zirconia and mixtures thereof. A catalyst comprising a zirconia refractory foam as carrier is specifically exemplified. The exemplified catalyst carriers have 390 and 1550 pores/cm² (50 and 100 ppi). This document clearly shows that the use of a ceramic foam having a higher number of pores in catalytic partial oxidation of a hydrocarbon feedstock results in an improved performance.

For successful operation on a commercial scale, the catalytic partial oxidation process must be able to

achieve a high conversion of the hydrocarbon feedstock at extreme high gas hourly space velocities, while the pressure drop over the catalyst bed should be low.

Further, the selectivity of the process to the desired products of carbon monoxide and hydrogen must be high.

Both these factors must be met using process equipment which is both economical to construct and economical to operate. In this respect, there exists a significant

problem in operating the catalytic partial oxidation process with a catalyst in a fixed bed arrangement having a high number of pores, in that the pressure drop

encountered when using the fixed bed is too high, and does not allow the process to operate under the high gas space velocities demanded in commercial operation. In

order to keep the pressure drop through the bed acceptably low at the very high gas space velocities

required, a very thin fixed bed of catalyst, for example a very thin layer of ceramic foam, is required. However, the mechanical strength of very thin monolithic

structures is insufficient to withstand the pressure differences encountered when operating at commercial acceptable conditions, while also handling might become difficult.

Surprisingly, it has now been found that the pressure drop over the catalyst bed at very high gas space

velocities can be significantly reduced, while still achieving a commercially acceptable rate of conversion and yield, when a specific monolithic structure is used.

In particular, it has been found that the pressure drop over the catalyst bed is significantly reduced and the catalyst bed has sufficient strength if a multiple layer monolithic structure is used comprising at least a first layer of porous material having relative small pores and

a second layer of porous material having relative large pores.

Accordingly, the present invention relates to a multiple layer monolithic structure comprising at least a first layer of porous material having a relatively high number of pores per cm and a second layer of porous material having a relatively low number of pores per cm. Reference herein to a monolithic structure is to any single porous material unit, suitably to any single porous refractory material unit, in which the pores constitute straight or tortuous, parallel or random elongate channels extending through the unit structure, i.e. having interconnected open-porosity. Reference herein to pores is to the openings or spaces between adjacent portions or lands of the monolithic structure. Thus, it will be appreciated that the pores referred to in respect of the present invention have a nominal diameter of the order of magnitude of 0.05 to 5 mm. These are to be contrasted with the micropores which may be present in the catalyst support material itself. Reference herein to the number of pores per cm is to the number of pores counted alongside a bar of 1 cm which is laid on the monolithic structure. A preferred method of measuring this is by using an enlarged photograph. In case of an uneven distribution of pores over the monolithic structure, reference herein to number of pores is to the number of pores averaged over different directions. The first layer of the multiple layer monolithic structure of the invention is suitably situated at the upstream end of the monolithic structure.

The monolithic structure suitably has a void fraction in the range of 0.4 to 0.95, preferably in the range of 0.6 to 0.9.

5 The monolithic structure of the present invention is particularly advantageous in that its use in catalytic conversion processes allows the application of very high gas hourly space velocities without causing a significant pressure drop, while providing sufficient strength to the catalyst.

10 The monolithic structure of the present invention is porous, as hereinbefore defined. The first layer of the multiple layer monolithic structure has preferably a high number of pores and has a higher number of pores per cm than the second layer. Thus, it will be appreciated that the first layer has smaller pores than the second layer. The first layer suitably contains at least 15 pores per cm (38 ppi), in particular at least 20 pores per cm (50 ppi). Preferred monolithic structures are those with 15 a first layer having a number of pores per cm in the range of from 20 to 120 (50 to 305 ppi), more preferably in the range of from 25 to 100 pores per cm (63 to 254 ppi). The second layer may have any number of pores per cm which is lower than that of the first layer, 20 preferably in the range of from 1 to 40 pores per cm (2 to 102 ppi), more preferably in the range of from 5 to 15 pores per cm (12 to 38 ppi).

25 The minimum thickness of the first layer of the monolithic structure is suitably defined by the thickness necessary to obtain sufficient catalytic conversion in the process in which it is used. The thickness thus depends on the intended use. Suitably the first layer has a thickness in the range of from 1 to 30 mm, preferably 30 in the range of from 1 to 20 mm, more preferably in the range of from 2 to 10 mm. The second layer may have a thickness in the range of from 1 to 10 cm, preferably in the range of from 1 to 5 cm. Preferably the thickness of each layer is equal over its whole width.

The first and second layer are adjacent layers. Any third or fourth or higher layer may have more or less pores per cm than the second layer. For example, in the case of a three layered monolith, the third layer may for example have a number of pores per cm in the same range as the first layer, or a number of pores per cm in the same range as the second layer.

In a particular aspect of the invention, the monolithic structure comprises an infinite number of very thin layers having a decreasing number of pores in downstream direction, thus creating a gradient in number of pores in that direction.

In the plane perpendicular to the central longitudinal axis, the multiple layer monolithic structure may have any suitable geometry. Various geometries are possible, for example square, rectangular or polygonal, but a round geometry is preferred. The width, or diameter in the case of the preferred round geometry, of the multiple layer monolithic structure may be of the order of from 1 cm to 1 m or more in the largest dimension. Preferably the largest dimension is in the range of from 1 to 50 cm, more preferably in the range of from 5 to 30 cm.

The multiple layer monolithic structure may have any shape. Preferably the first layer is situated at the upstream end of the monolithic structure. In a preferred aspect of the invention the monolithic structure has the shape of a truncated cone. Suitably, the downstream end of the monolithic structure is co-planar with the upstream end. In one embodiment of the invention, the multiple layer monolithic structure has the shape of a curved co-planar truncated cone. In an alternative aspect of the invention the monolithic structure comprises a curved upstream end and a flat downstream end. Suitably

the monolithic structure is defined by a cone half angle in the range of from 0° to 20° to the axis, preferably in the range of from 5° to 15°. Reference herein to the cone half angle is to the angle defined by the cone central longitudinal axis and any "generator"; i.e. line
5 contained in the cone surface thereof.

The porous material of the multiple layer monolithic structure of the present invention suitably is an inorganic material of high temperature resistance, in
10 particular refractory oxides, such as silica, alumina, titania, zirconia and mixtures thereof. The choice of material will generally depend on the intended use. Preferably the porous material of the monolithic structure is zirconia-based. Any suitable zirconia-based
15 material may be employed. The material preferably comprises at least 70% by weight zirconia, for example selected from known forms of (partially) stabilised zirconia or substantially pure zirconia. (Partially) stabilised zirconia comprising oxides of one or more of
20 the rare earth, Group IIIB or Group IIA elements of the Periodic Table of the Elements are particularly preferred zirconia-based materials. References herein to the Periodic Table of the Elements are to the CAS version, as published in the CRC Handbook of Chemistry and Physics,
25 68th Edition. Most preferred zirconia-based materials comprise zirconia stabilised or partially-stabilised by one or more oxides of Mg, Ca, Al, Y, La or Ce.

The porous material of the monolithic structure may be an inorganic material as above described coated with a
30 further material having a desirable property rendering enhanced thermal shock resistance (for example SiC, SiO₂, Al₂O₃ or a mixture thereof), enhanced catalyst stability or enhanced thermal stability.

The monolithic structure may have any suitable form. One form of monolithic structure is that of an extruded honeycomb. Extruded honeycomb materials are characterised by having a plurality of straight, elongate, parallel channels extending through the structure.

Preferred monolithic structures are those having a high tortuosity. The term "tortuosity" is a common term in the art which, as used herein, is defined as the ratio of the length of the path followed by a gas flowing through the structure to the length of the shortest possible straight line path through the structure. Thus, it follows that the extruded honeycomb structures have a tortuosity of 1.0. The monolithic structure of the present invention preferably has high tortuosity, that is a tortuosity of greater than 1.1. The monolithic structure preferably has a tortuosity in the range of from 1.1 to about 10.0, more preferably in the range of from 1.1 to about 5.0. A most suitable range of tortuosity for the monolithic structure is from 1.3 to 4.0.

A most suitable monolithic structure is a ceramic foam. A multiple layer ceramic foam may be prepared by any suitable method. One suitable preparation method is combining two or more layers of a flexible open-pore organic polymer foam, e.g. polyurethane foam, which are impregnated with ceramic material, followed by calcining at temperatures above 1000 °C in order to remove the organic precursor and to cause the ceramic to sinter. Alternatively, two or more layers of organic polymer foam may be combined prior to impregnation with ceramic material. The combined layers are then impregnated with ceramic material and calcined. Another possibility is to combine two or more layers of ceramic foam to form one multiple layer ceramic foam.

The multiple layer monolithic structure according to the present invention may contain more than two layers, but most suitably contains one first layer as hereinbefore defined and one second layer as hereinbefore defined, thus constituting a dual layer monolithic structure.

The present invention also relates to a catalyst or a catalyst precursor comprising a catalytically active material or a precursor therefor supported on a multiple layer monolithic structure as hereinbefore defined. In this respect, the term "catalytically active material" is a reference to such components in the broadest sense, that is including components which exhibit catalytic activity per se, together with other components which, acting as promoters, stabilisers and the like, have a beneficial effect on the catalytic performance of the components present.

The whole multiple layer monolithic structure may have been provided with a catalytically active material or a precursor therefor. In a specific embodiment of the present invention only the first layer of the multiple layer monolithic structure has been provided with a catalytically active material or a precursor therefor.

The catalytically active material to be supported on the multiple layer monolithic structure may be selected from any suitable component or combination of components known in the art. The selection of catalytically active components or combination of components will depend upon the intended end use of the catalyst.

One process for which the catalyst of the present invention is particularly suitable is the catalytic partial oxidation of a hydrocarbonaceous feed to synthesis gas. Suitable catalytically active components for the catalytic partial oxidation process are the

Group VIII elements. Preferred elements for the catalytic partial oxidation process are rhodium, platinum, palladium, osmium, iridium and ruthenium, more preferred elements are rhodium and iridium. A further application of the catalyst of the present invention is in a process for the reduction of nitric oxides. Suitable catalytically active components for the reduction of nitric oxides include vanadium, titanium and a mixture thereof. The multiple layer monolithic structure of this invention is also suitable for use in the manufacture of ethylene oxide. A most suitable catalytically active component for this application is silver, especially in combination with one or more promoters.

The catalyst or the catalyst precursor of the present invention may be prepared by processes known in the art. A most suitable process is the impregnation of the multiple layer monolithic structure with the catalytically active material or a precursor thereof. Impregnation typically comprises contacting the monolithic structure with a solution of a compound of the catalytically active material or precursor thereof, followed by drying and, optionally, calcining the resulting material.

The catalyst may comprise the catalytically active material in any suitable amount to achieve the required level of activity. Typically, the catalyst comprises the active metal in an amount in the range of from 0.01 to 20% by weight, preferably from 0.02 to 10% by weight, more preferably from 0.1 to 7.5% by weight.

From the foregoing it will be apparent that the multiple layer monolithic structure of the invention is admirably suited for the use as a catalytic support in catalytic processes wherein high space velocities are required. In particular it will be apparent that the

monolithic structure enables operation under very high space velocities without causing a significant pressure drop.

5 Accordingly the present invention further relates to a process for the catalytic conversion of fluid substrate comprising contacting the substrate with a catalyst as hereinbefore defined. Reference herein to fluid is to a gaseous or liquid fluid, preferably a gaseous fluid, or a combination thereof.

10 Preferably the process of the invention is a process for the catalytic partial oxidation of a hydrocarbon feedstock, which comprises contacting a feed comprising a hydrocarbon feedstock and an oxygen-containing gas with a catalyst as hereinbefore defined at a pressure in the
15 range of up to 150 bara, preferably from 2 to 125 bara, more preferably from 5 to 100 bara, at a temperature in the range of from 750 to 1400 °C, more preferably in the range of from 850 to 1300 °C, and at a gas hourly space velocity in the range of from 20,000 to
20 100,000,000 Nl/kg/hr, preferably in the range of from 50,000 to 50,000,000 Nl/kg/hr, more preferably from 500,000 to 30,000,000 Nl/kg/hr. Reference herein to temperature is to the temperature of the fluid leaving the monolithic structure.

25 The preferred process is suitably used to prepare a mixture of carbon monoxide and hydrogen from any hydrocarbonaceous feedstock. This process is a means to obtain very useful products known in the art as synthesis gas by means of an exothermic reaction, by which the
30 molar ratio of carbon monoxide to hydrogen in the products may be controlled by means of choice of feedstock and operating conditions to give a desired molar ratio of carbon monoxide to hydrogen in the products.

The hydrocarbonaceous feedstock is in the gaseous phase when contacting the catalyst. The feedstock may contain compounds that are liquid and/or compounds that are gaseous under standard conditions of temperature and pressure (i.e. at 0 °C and 1 atm.). The process is particularly suitable for the partial oxidation of methane, natural gas, associated gas or other sources of light hydrocarbons. In this respect, the term "light hydrocarbons" is a reference to hydrocarbons having from 1 to 5 carbon atoms. The process may be advantageously applied in the conversion of gas from naturally occurring reserves of methane which contain substantial amounts of carbon dioxide. The feed preferably comprises methane in an amount of at least 50% by volume, more preferably at least 70% by volume, especially at least 80% by volume.

The hydrocarbonaceous feedstock is contacted with the catalyst as a mixture with an oxygen-containing gas. Air is suitable for use as the oxygen-containing gas. However, the use of oxygen-enriched air or pure oxygen as the oxygen-containing gas may be preferred. In this way, the need for handling a large volume of inert gas, for example nitrogen when using air as the oxygen-containing gas, is avoided. The feed may optionally comprise steam. Optionally, the feed may comprise carbon dioxide in a concentration of up to 10% by volume of the total feed mixture.

The hydrocarbonaceous feedstock and the oxygen-containing gas are preferably present in the feed in such amounts as to give an oxygen-to-carbon ratio in the range of from 0.3 to 0.8, more preferably, in the range of from 0.45 to 0.75. References herein to the oxygen-to-carbon ratio refer to the ratio of oxygen in the form of molecules (O₂) to carbon atoms present in the hydrocarbon feedstock. Oxygen-to-carbon ratios in the region of the

stoichiometric ratio of 0.5, that is ratios in the range of from 0.45 to 0.65, are especially preferred. If steam is present in the feed, the steam-to-carbon ratio is preferably in the range of from above 0.0 to 3.0, more preferably from above 0.0 to 2.0. The hydrocarbonaceous feedstock, the oxygen-containing gas and steam, if present, are preferably well mixed prior to being contacted with the catalyst. The feed mixture is preferably preheated prior to contacting the catalyst.

The feed is preferably contacted with the catalyst under adiabatic conditions. For the purposes of this specification, the term "adiabatic" is a reference to reaction conditions under which substantially all heat loss and radiation from the reaction zone are prevented, with the exception of heat leaving in the gaseous effluent stream of the reactor.

The invention is now illustrated in a non-limiting manner with reference to Figures 1 to 4, wherein Figures 1 to 4 are each a cross-section through the monolithic structure of the invention in the plane of the central longitudinal axis thereof.

In Figure 1a is shown a dual layer monolithic structure of the invention comprising a first layer (1) of porous material having a relatively high number of pores per cm, a second layer (2) of porous material having a relatively low number of pores per cm, an upstream end (3), and a downstream end (4). The monolithic structure in Figure 1b has the shape of a co-planar truncated cone. The cone half angle (5) is illustrated with central longitudinal axis A in the inset, selected from ranges as hereinbefore defined.

In Figures 2 to 4 are shown alternative embodiments of the dual layer monolithic structure of Figure 1. In Figure 2 a curved co-planar truncated cone is shown. In

Figure 3 is shown a severely curved co-planar structure. In Figure 4 is shown a multiple layer monolithic structure wherein the upstream cone end (3) is curved and the downstream end (4) is flat.

5 The invention will now be illustrated further by means of the following Examples.

EXAMPLE 1

1a. Ceramic foam (according to the invention)

10 Foam 1 is a dual layer ceramic foam consisting of zirconia, partially stabilised with 4.2 mol% yttrium (4.2% Y-PSZ). The foam has a cylindrical shape with a thickness of 20 mm and a diameter of 47 mm; the foam volume is 35 ml. The foam has a 10 mm thick first layer having 65 pores per inch (26 pores per cm) and a second
15 10 mm thick layer having 20 pores per inch (8 pores per cm).

1b. Pressure drop

20 The pressure drop of foam 1 was measured ten times (i.e. once on ten different pieces of foam). One piece of foam was loaded into a tube reactor with the first layer at the feed inlet side. Nitrogen was fed to the reactor at an inlet velocity of 5 m/s. The resulting pressure drop over the foam was measured.

The average pressure drop is given in Table 1.

25 1c. Preparation of catalyst

30 Foam 1 was several times impregnated with a solution containing iridium chloride and zirconium nitrate followed by drying until the foam comprised 5% wt iridium and 7% wt zirconium based on ceramic foam (4.2% Y-PSZ). The impregnated foam was then dried at 120 °C for 4 hours and calcined at 700 °C for 4 hours.

1d. Catalytic partial oxidation

A foam catalyst prepared as hereinbefore described under 1c was loaded into a tube reactor. Methane

(12,000 Nl/h) was thoroughly mixed with air and preheated to a temperature of 150-200 °C. The resulting oxygen-to-carbon ratio was 0.65. The mixture of methane and air was fed to the reactor at a pressure of 11 bara. The methane conversion, CO selectivity and temperature of the gas leaving the catalyst were measured. The process parameters and the results are given in Table 1.

EXAMPLE 2 (not according to the invention)

2a. Ceramic foams (not according to the invention)

Foam 2 is a single layer ceramic foam consisting of 4.2% Y-PSZ. The foam has the same dimensions as foam 1 (thickness: 20 mm; diameter: 47 mm) and has 65 pore per inch (26 pores per cm).

Foam 3 is a single layer ceramic foam consisting of 4.2% Y-PSZ. The foam has the same dimensions as foam 1 (thickness: 20 mm; diameter: 47 mm) and has 20 pores per inch (8 pores per cm).

2b. Pressure drop

The pressure drops of foam 2 and of foam 3 were measured as described above for foam 1.

2c. Preparation of catalyst

Catalysts were prepared from foam 2 and from foam 3 as described under 1c. The resulting catalysts comprised 7% wt Zr and 5% wt Ir on ceramic foam (4.2% Y-PSZ).

2d. Catalytic partial oxidation

Catalytic partial oxidation was performed with each of the foam catalysts. The catalytic partial oxidation was performed as described under 1d. The results are given in Table 1.

TABLE 1

| | EXAMPLE 1 | EXAMPLE 2 | |
|--------------------------|----------------------------------|---------------------|---------------------|
| | foam 1 | foam 2 | foam 3 |
| Foam properties | | | |
| thickness (mm) | 10 ^a /10 ^b | 20 | 20 |
| porosity (ppi) | 65 ^a /20 ^b | 65 | 20 |
| Pressure drop (mbar) | 50 | 150 | <10 |
| CPO process parameters | | | |
| pressure (bara) | 11 | 11 | 11 |
| GHSV (Nl/l/h) | 1.4 10 ⁶ | 1.4 10 ⁶ | 1.4 10 ⁶ |
| O ₂ :C | 0.65 | 0.65 | 0.65 |
| CPO process performance | | | |
| methane conversion (%wt) | 85 | 90 | 77 |
| CO selectivity (%wt) | 85 | 86 | 82 |
| T syngas (°C) | 960 | 920 | 1085 |

^a first layer

^b second layer

C L A I M S

1. A multiple layer monolithic structure comprising at least a first layer of porous material having a relatively high number of pores per cm and a second layer of porous material having a relatively low number of pores per cm.

2. A multiple layer monolithic structure according to claim 1, wherein the first layer has a number of pores per cm in the range of from 20 to 120 (50 to 305 ppi), preferably in the range of from 25 to 100 pores per cm (63 to 254 ppi), and the second layer has a number of pores per cm in the range of from 1 to 40 (2 to 102 ppi), preferably in the range of from 5 to 15 pores per cm (12 to 38 ppi).

3. A multiple layer monolithic structure according to claim 1 or 2, wherein the first layer has a thickness in the range of from 1 to 20 mm, preferably in the range of from 2 to 10 mm and the second layer has a thickness in the range of from 1 to 5 cm.

4. A multiple layer monolithic structure according to any of claims 1 to 3, which has a round geometry in the plane perpendicular to the central longitudinal axis, preferably has the shape of a truncated cone, more preferably a co-planar truncated cone, preferably with a diameter in the range of from 1 to 50 cm, more preferably in the range of from 5 to 30 cm.

5. A multiple layer monolithic structure according to any of claims 1 to 4, wherein the porous material is an inorganic material, preferably a refractory oxide, more preferably silica, alumina, titania, zirconia or a

mixture thereof, most preferably zirconia or a stabilised zirconia.

5 6. A multiple layer monolithic structure according to any of claims 1 to 5, wherein at least one layer of the monolithic structure is a ceramic foam, preferably all layers are a ceramic foam.

7. A multiple layer monolithic structure according to any of claims 1 to 6, which is a dual layer monolithic structure.

10 8. A catalyst or a catalyst precursor comprising a multiple layer monolithic structure according to any of claims 1 to 7, wherein at least the first layer of the monolithic structure has been provided with a catalytically active material or a precursor therefor,
15 preferably a metal or metal compound derived from one or more of the elements of group VIII of the Periodic Table of the Elements, more preferably rhodium, platinum, palladium, osmium, iridium or ruthenium or a compound derived therefrom, even more preferably rhodium or
20 iridium or a compound derived therefrom.

9. A process for the catalytic conversion of a fluid substrate comprising contacting the substrate with a catalyst according to claim 8.

25 10. A process according to claim 9, which is a process for the catalytic partial oxidation of a hydrocarbonaceous feedstock, which comprises contacting a feed comprising a hydrocarbonaceous feedstock and an oxygen-containing gas in amounts giving an oxygen-to-carbon ratio of from 0.3 to 0.8, preferably from 0.45 to 0.75
30 with a catalyst according to claim 8 at a pressure of up to 150 bara, preferably in the range of from 2 to 125 bara, more preferably from 5 to 100 bara, at a temperature in the range of from 750 to 1400 °C, preferably from 850 to 1300 °C, and at a gas hourly space velocity

in the range of from 20,000 to 100,000,000 Nl/kg/hr,
preferably from 50,000 to 50,000,000 Nl/kg/hr, more
preferably from 500,000 to 30,000,000 Nl/kg/hr,
preferably under substantially adiabatic conditions.

Fig.1a.

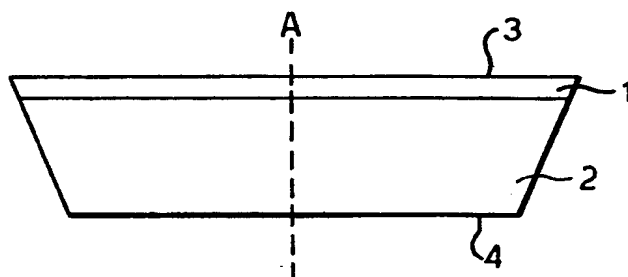


Fig.2.

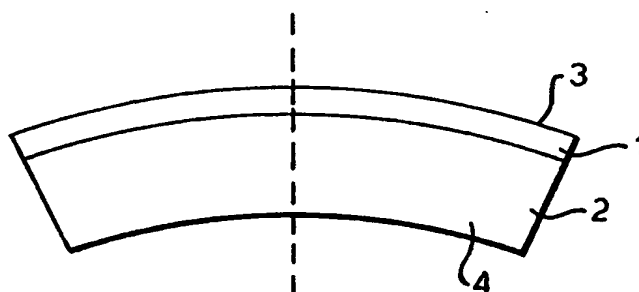


Fig.1b.

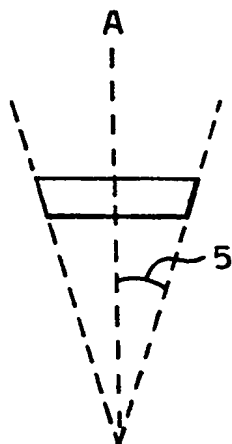


Fig.3.

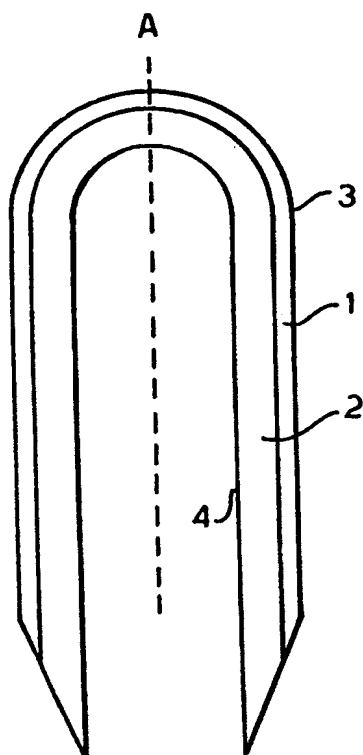
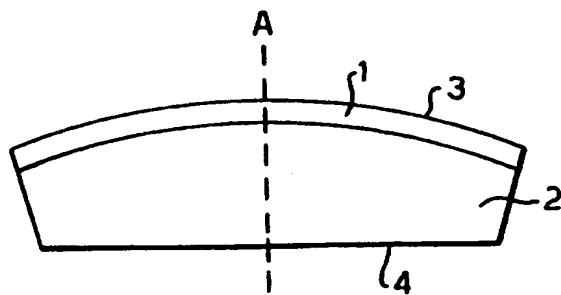


Fig.4.



INTERNATIONAL SEARCH REPORT

International Application No.
PCT/EP 98/08547

| | | |
|---|---|-----------------------|
| A. CLASSIFICATION OF SUBJECT MATTER IPC 6 B01J35/04 C04B38/00 B01J35/10 B01J37/02 | | |
| According to International Patent Classification (IPC) or to both national classification and IPC | | |
| B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) IPC 6 B01J C04B C01B | | |
| Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched | | |
| Electronic data base consulted during the international search (name of data base and, where practical, search terms used) | | |
| C. DOCUMENTS CONSIDERED TO BE RELEVANT | | |
| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
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| <div style="display: flex; justify-content: space-between;"> <input checked="" type="checkbox"/> Further documents are listed in the continuation of box C. <input checked="" type="checkbox"/> Patent family members are listed in annex. </div> | | |
| * Special categories of cited documents : | | |
| <div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&" document member of the same patent family</p> </div> </div> | | |
| Date of the actual completion of the international search <div style="text-align: center;">27 May 1999</div> | Date of mailing of the international search report <div style="text-align: center;">07/06/1999</div> | |
| Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016 | Authorized officer <div style="text-align: center;">Thion, M</div> | |

INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
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